Thick Low-Cost, High-Power Lithium-Ion Electrodes via Aqueous Processing

OAK RIDGE National Laboratory | National Transportation Research Center

PI: Jianlin Li; Georgios Polyzos; Zhijia Du

2022 DOE Annual Merit Review, Project ID = BAT164

Timeline
- Start: October 1, 2014
- End: September 30, 2022
- Percent Complete: 90%

Budget
- FY22 - $450K

Barriers
- Reduce EV batter-pack cost to $80-100/kWh
- Advance Li-ion HEV/PHEV battery systems with low-cost electrode architectures
- Achieve deep discharge cycling target of 1000 cycles for EV 2022

RELEVANCE

- Correlate ***aqueous*** colloidal dispersion properties and ***thick*** electrode coatings to cell performance to reduce LIB pack cost.
- Demonstrate an energy density to >300 Wh/kg (cell level) for solid state batteries.

OBJECTIVES

Main Objective: To improve cell energy and power density and reduce battery pack cost by tailored electrode architecture via aqueous processing and utilizing high energy high voltage cathode materials.

- Fabricate thick and crack-free composite cathodes via aqueous processing
- Evaluate compatibility of high Ni-NMC with aqueous processing
- Develop bilayer electrodes via freeze tape casting for improved rate performance
- Develop polymer electrolyte for solid-state batteries
- Understand the conduction mechanism in polymer electrolyte and optimize the formulation
- Demonstrate a solid-state battery with an energy density ≥300 Wh/kg (cell level).

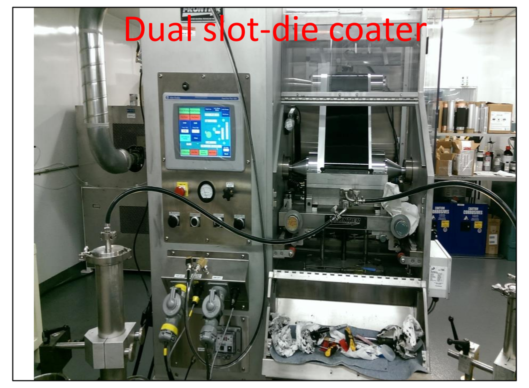
PROJECT MILESTONES

Status	SMART Milestones	Description
3/31/22 Completed	Quarterly Progress Measure (Regular)	Demonstrate high energy and power density from layered electrodes and freeze tape cast, achieving >10% improvement at high-rate performance
6/30/22 On track	Annual (Stretch)	Fabrication composite cathodes for solid-state batteries and demonstrate an energy density > 300 Wh/kg at cell level and > 20 cycles

TECHNICAL APPROACH

Problems:
- Electrode cracking in thick electrodes
- Mass transport limitations thick electrodes

Technical approach and strategy:
- ❑ Fabricate crack-free NMC622 electrodes with high areal loading (4-8 mAh/cm^2) via aqueous processing
- ❑ Develop bilayered electrode architecture via freeze tape casting
- ❑ Develop composite polymer electrolyte
- ❑ Characterize the electrochemical performance of the composite electrolytes
- ❑ Study the electrolyte-cathode interface
- ❑ Evaluate rate performance and long term cyclability



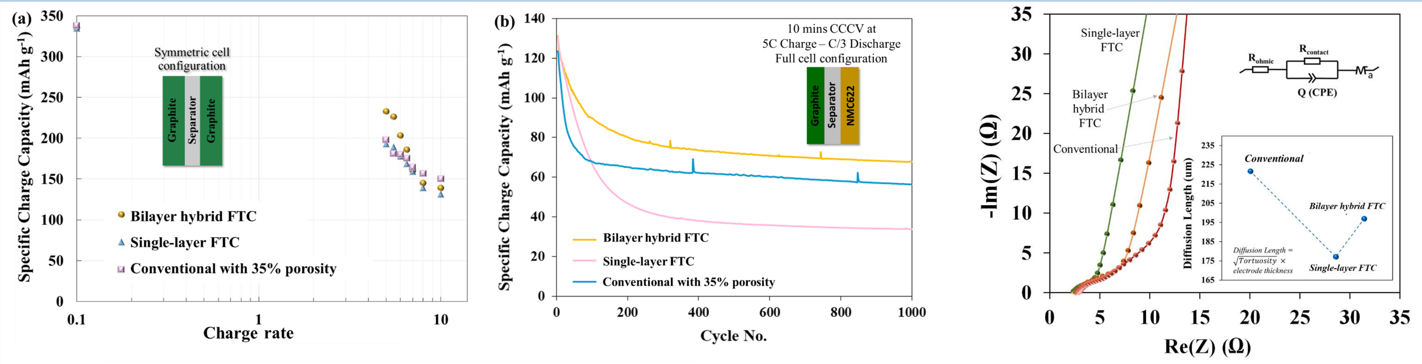
CALLABORATION

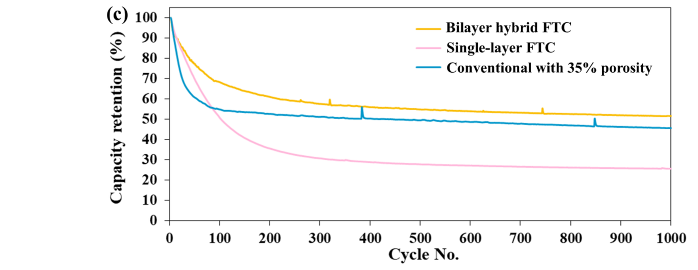
Partners
- National Labs: Argonne National Laboratory, Idaho National Laboratory
- Battery Manufacturers: Navitas Systems
- Active Material Suppliers: Targray, Superior Graphite, Forge Nano
- Inactive Material Suppliers: JSR Micro, Solvay Specialty Polymers, Ashland, IMERYS
- Equipment/Coating Suppliers: PPG Industries, Frontier Industrial Technology, B&W MEGTEC, DataPhysics
- Universities: KIT, Binghamton University, University of Picardy Jules Verne

TECHNICAL ACCOMPLISHMENTS

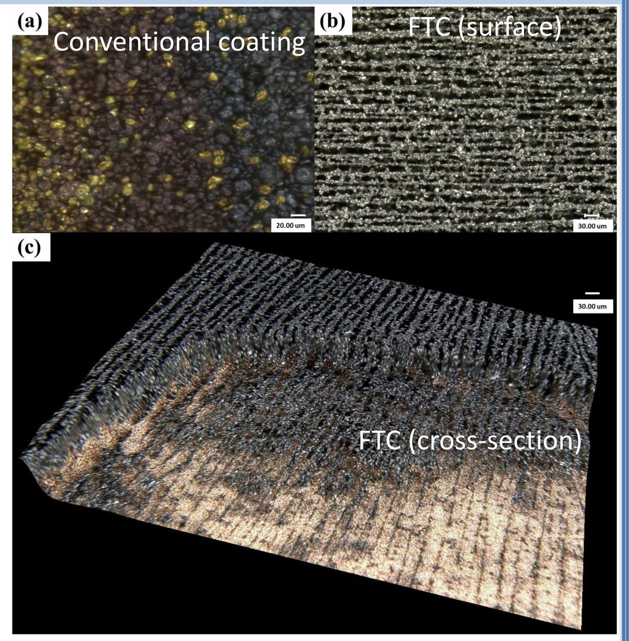
Executive Summary:
- ❑ Demonstrated crack- and defect-free flexible graphite and NMC622 cathodes by freeze tape casting (FTC)
- ❑ Tailored the aqueous slurry formulations to control the structure of the cathode
- ❑ Demonstrated 20% improvement in capacity under 5C and 10 min charging from graphite anodes
- ❑ Delineated the Li$^+$ ion conductivity mechanism in hybrid solid state electrolytes based on Al-LLZO
- ❑ Study the polymer electrolyte Al-LLZO interface
- ❑ Correlated the conductivity mechanism with the macroscopic electrochemical performance
- ❑ Investigated the electrochemical stability of LiTFSI vs LiFSI
- ❑ Designed principles to synthesize electrolytes with tailored properties

Developed bilayer graphite anode vis FTC and demonstrated ~20% improvement in capacity

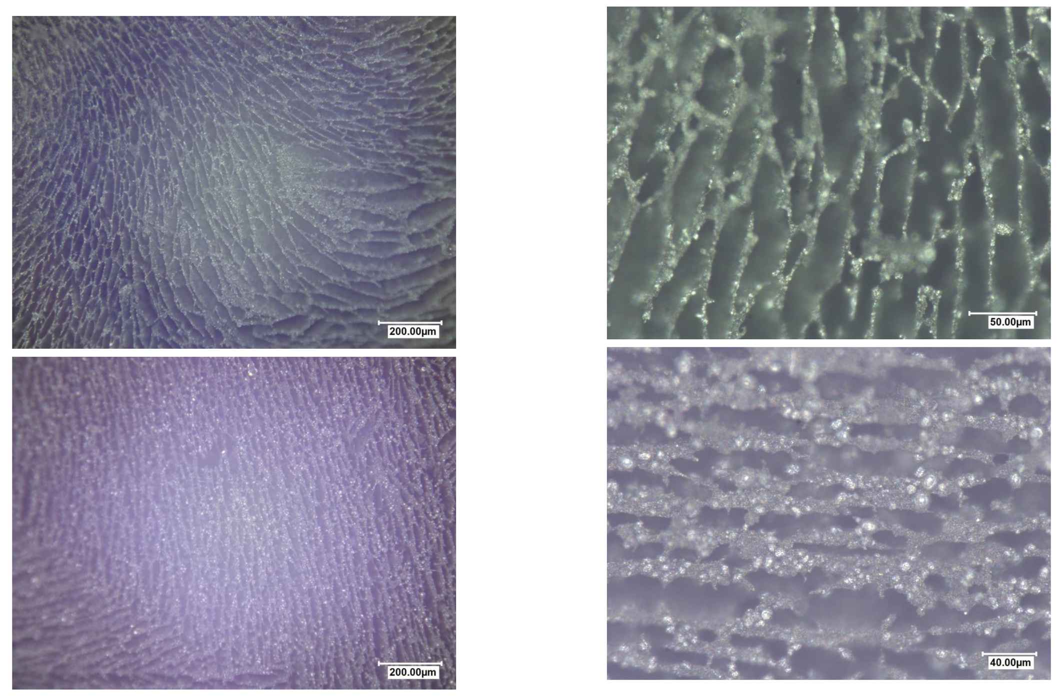




The bilayer graphite anode demonstrated excellent mechanical integrity, shorter diffusion length, ~20% improvement in capacity under 5C charging and 10 min total charging time, and improved cycle life.

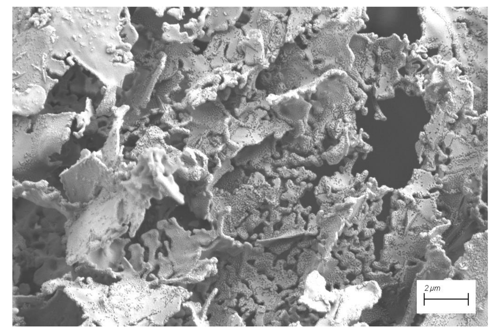


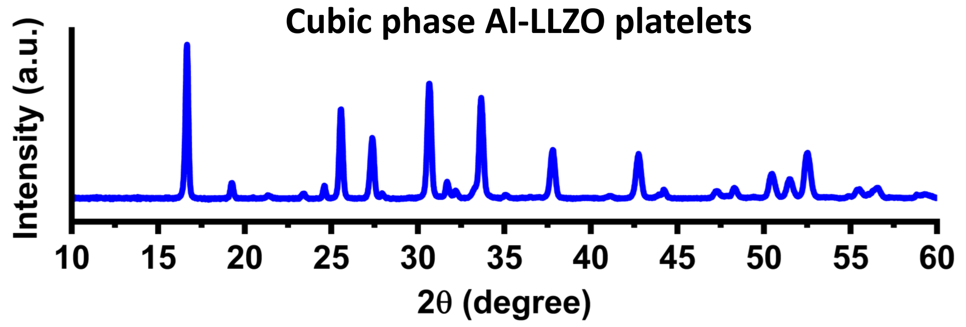
Demonstrated crack-free and thick NMC622 cathodes (4.7 mAh/cm^2) via FTC

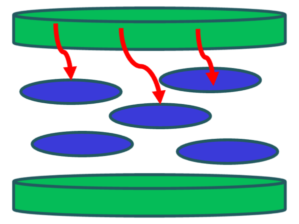


- **Vertical channels were formed in the NMC622 cathode**
- **Large void space in the channels**
- **Denser and more durable structured cathodes enabled by tailoring the water content of the slurry formulation**

Composite electrolytes based on poly(ethylene oxide) filled with Al–doped $Li_7La_3Zr_2O_{12}$ platelets



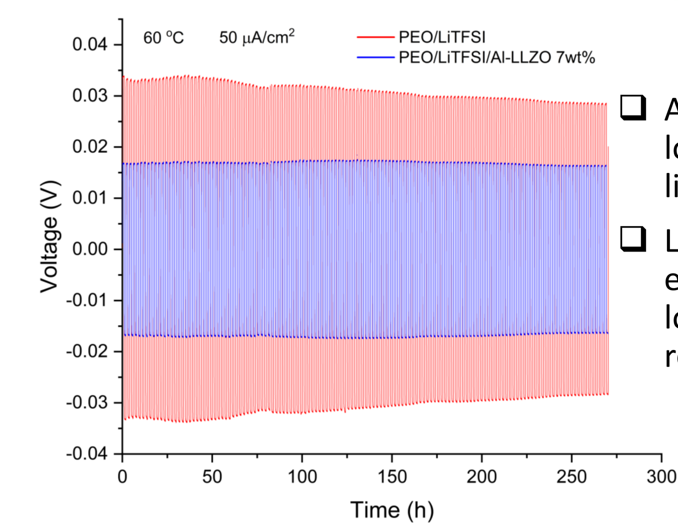




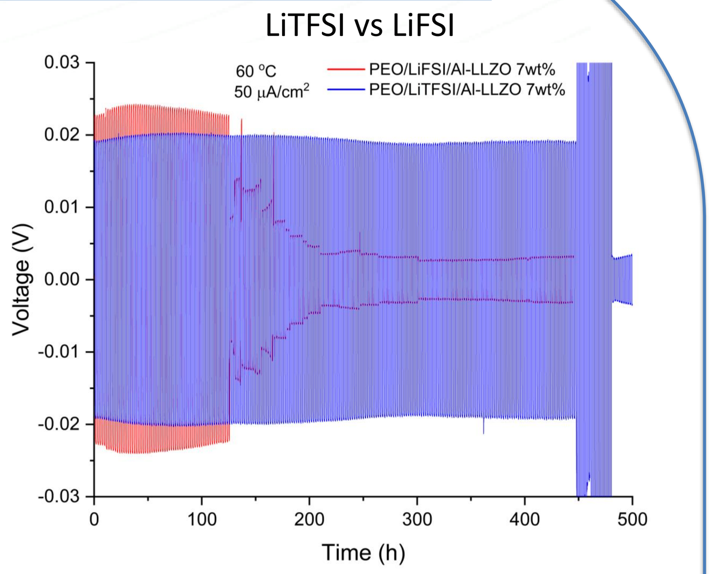
Platelet Al-LLZO
- ❑ **Better against dendrite formation**
- ❑ **Good adsorption of the polymer electrolyte**

Optimize formulation and processing conditions
- ❑ **Li salts: LiTFSI, LiFSI**
- ❑ **PEO, LiFTSI, LiFSI dissolved in acetonitrile (ACN) or other solvents**

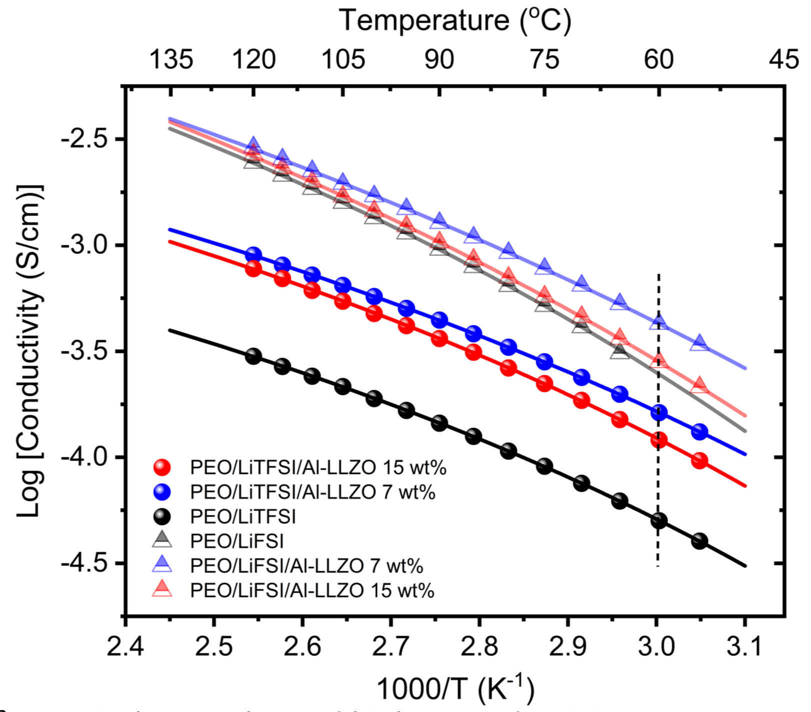
Addition of Al-LLZO increased conductivity and reduced polarization



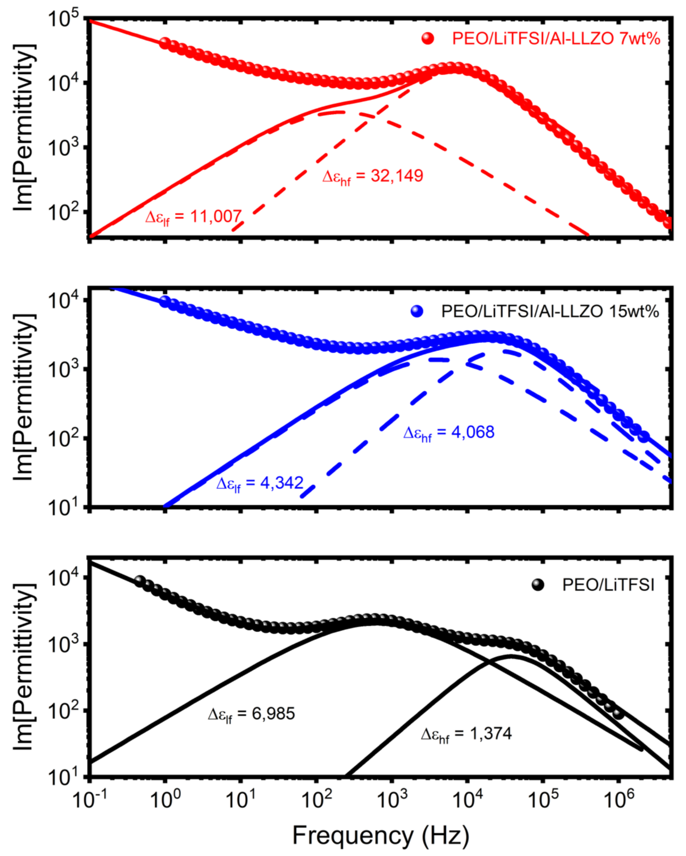
- ❑ Addition of Al-LLZO has lower overpotential in lithium plating/stripping
- ❑ LiTFSI based composite electrolyte has significantly longer life than the respective LiFSI electrolyte



Addition of Al-LLZO accelerated lithium ion dynamics



bimodal mechanism: Strongly and loosely coordinated ions



- ❑ 7wt% Al-LLZO showed higher conductivity
- ❑ The electrolyte with LiFSI showed lower glass transition temperature
- ❑ High frequency relaxation more significant with Al-LLZO indicating faster lithium ion dynamics

SUMMARY

- ❑ Fabricated thick and crack-free NMC622 cathodes (4.7 mAh/cm^2) via FTC with tailored structure
- ❑ Demonstrated 20% improvement in fast charge capability in bilayer graphite anode via FTC
- ❑ The platelet Al-LLZO fillers resulted in substantial improvement of the stripping/platting performance
- ❑ The addition of AL-LLZO dramatically increased conductivity with faster dynamics
- ❑ LiFSI salt showed higher conductivity than LiTFSI in composite electrolyte
- ❑ The Al-LLZO fillers significantly change the conductivity mechanism: The Li ion dynamics become faster and the conductivity increases: Possible interfacial conductivity mechanism
- ❑ The ratio of the dielectric strengths (number density of the ions contributing to the mechanism) is associated with the electrolyte conductivity

FUTURE WORK

- ❑ Test the FTC cathodes
- ❑ Characterize the chemical structure in the nanoscale of the polymer electrolytes using XPS techniques
- ❑ Characterize the electrolyte structure using SAXS techniques
- ❑ Fabricate solid state batteries and test the electrochemical performance

Any proposed future work is subject to change based on funding levels

ACKNOWLEDGEMENTS

- U.S. DOE EERE VTO (David Howell, Peter Faguy, Haiyan Croft)
- ORNL contributors
  - Georgios Polyzos
  - Dhrupad Parikh
  - Ronnie Tao
  - Alexander Kukay
  - Yangyang Wang
  - Blake Hawley
  - Anand Parejiya
  - Ritu Sahore
  - Zhijia Du
  - Ilias Belharouak
- External Technical Collaborators:
  - Ozge Kahvecioglu Ferdun
  - Congrui Jin
  - Alejandro Franco
  - Abbos Shodiev
  - Xiangbo Meng
  - James Banas
  - Gregg Lytle
  - Shufu Peng